Resolving Ultrafast Dynamics With X-Rays

Moses Marsh
Shpyrko Group
2/4/11

How fast?

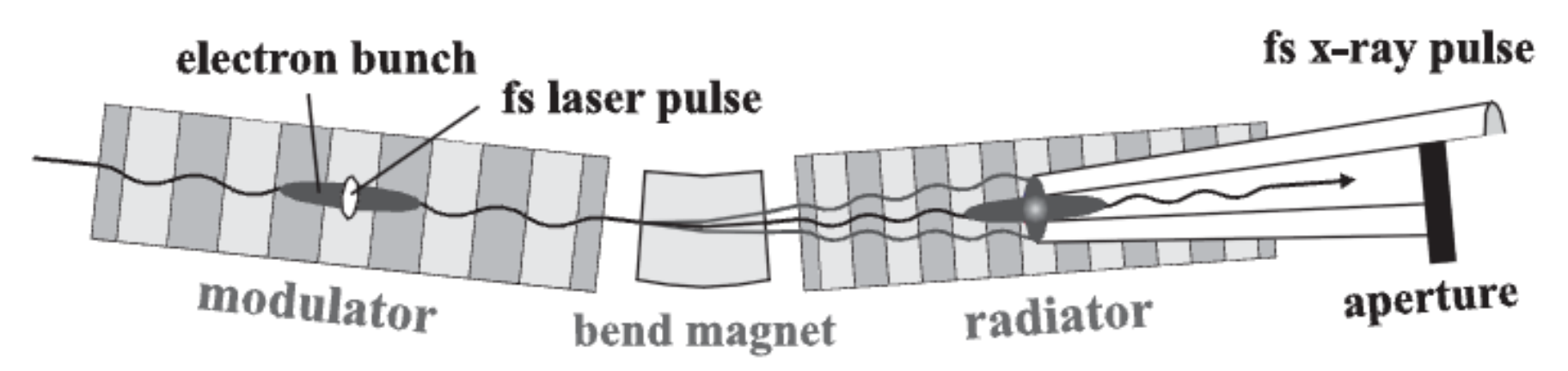
- Time resolution of ~100 femtoseconds is necessary to measure atomic motion in lattice vibrations (phonons), melting , and solid-solid phase transitions
- This is also roughly the timescale of the transfer of energy from excited electrons to the lattice (thermalization)
- Electron excitations happen on the order of fs

X-ray Source Timescales

- Synchrotron
  - ~100 ps x-ray pulse, one pulse every ~150 ns (APS)
- Free electron laser (FEL)
  - ~100 fs x-ray pulse, one pulse every 8.3 ms (LCLS)
- Pump-probe measurements
  - ~100 fs IR laser pulse
  - Synchronization within a few ps of the pulses is accomplished by “locking in” the repetition rates

Shorter Pulses: Slicing

- A ~50fs IR laser pulse modulates the energy of a “slice” of the electron bunch, which is then spatially separated by a bending magnet.



Khan, et al 2006, BESSY II

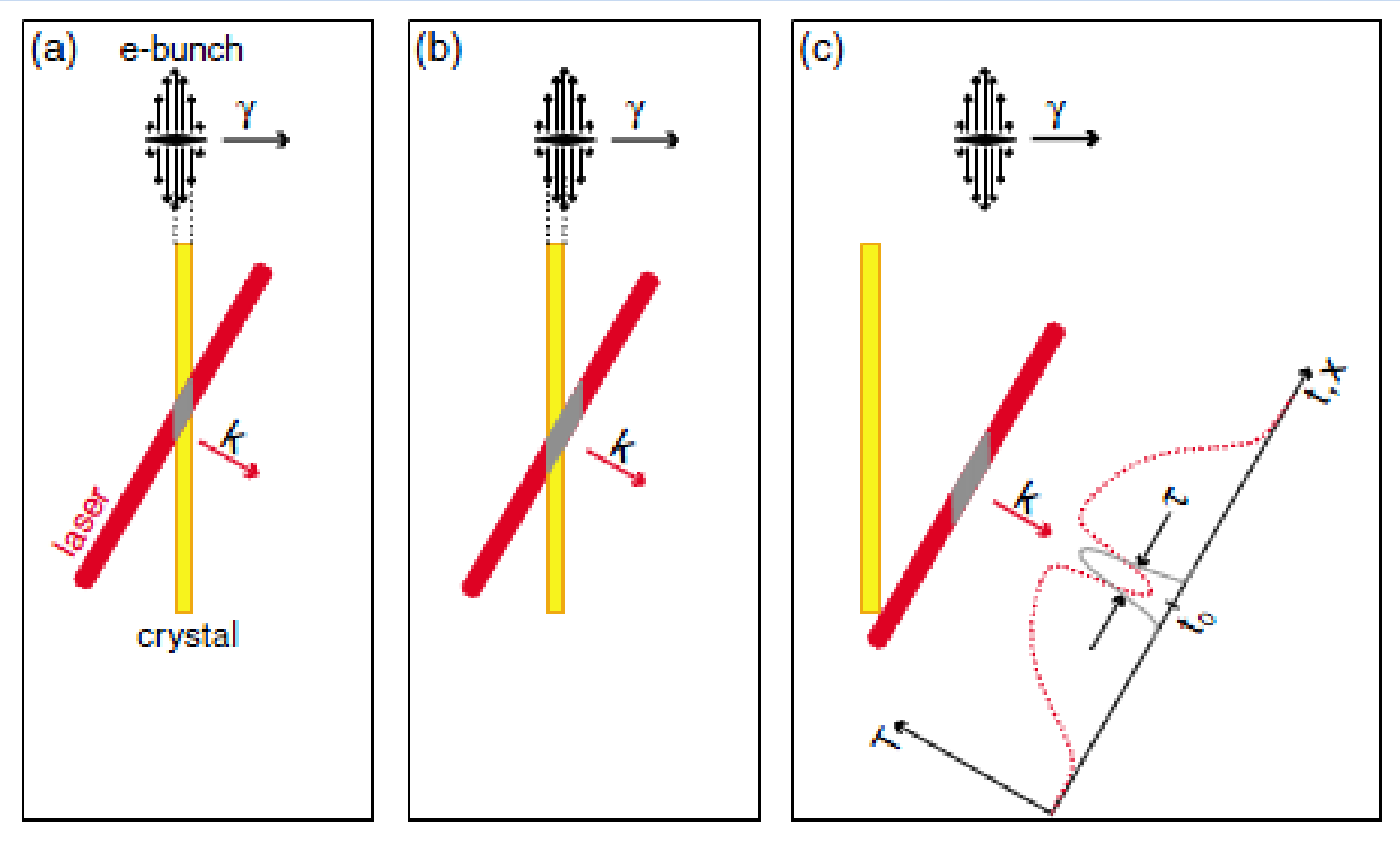
Shorter Pulses: Slicing

- The resulting x-ray pulse is ~100fs
- The pulse intensity is 10^{-4} of the total radiation
- Intensity can be improved by using a higher repetition rate for the laser

Pump-probe Synchronization: Electro-Optic Sampling

- A crystal (e.g. ZnTe) is placed near the electron bunch as it passes through the accelerator.
- The large electric field of the bunch produces a temporary change in the index of refraction of the crystal.
- An IR laser passing through the crystal will have its polarization changed as the bunch passes

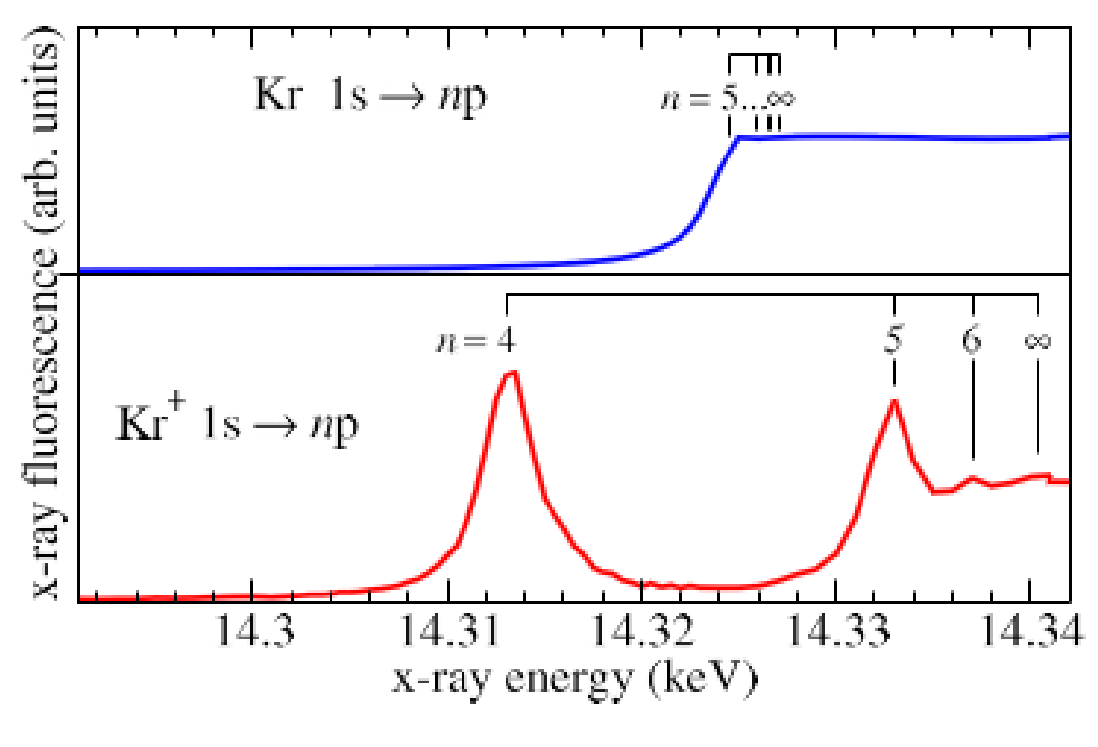
Pump-probe Synchronization: Electro-Optic Sampling



Cavalieri, et al 2005

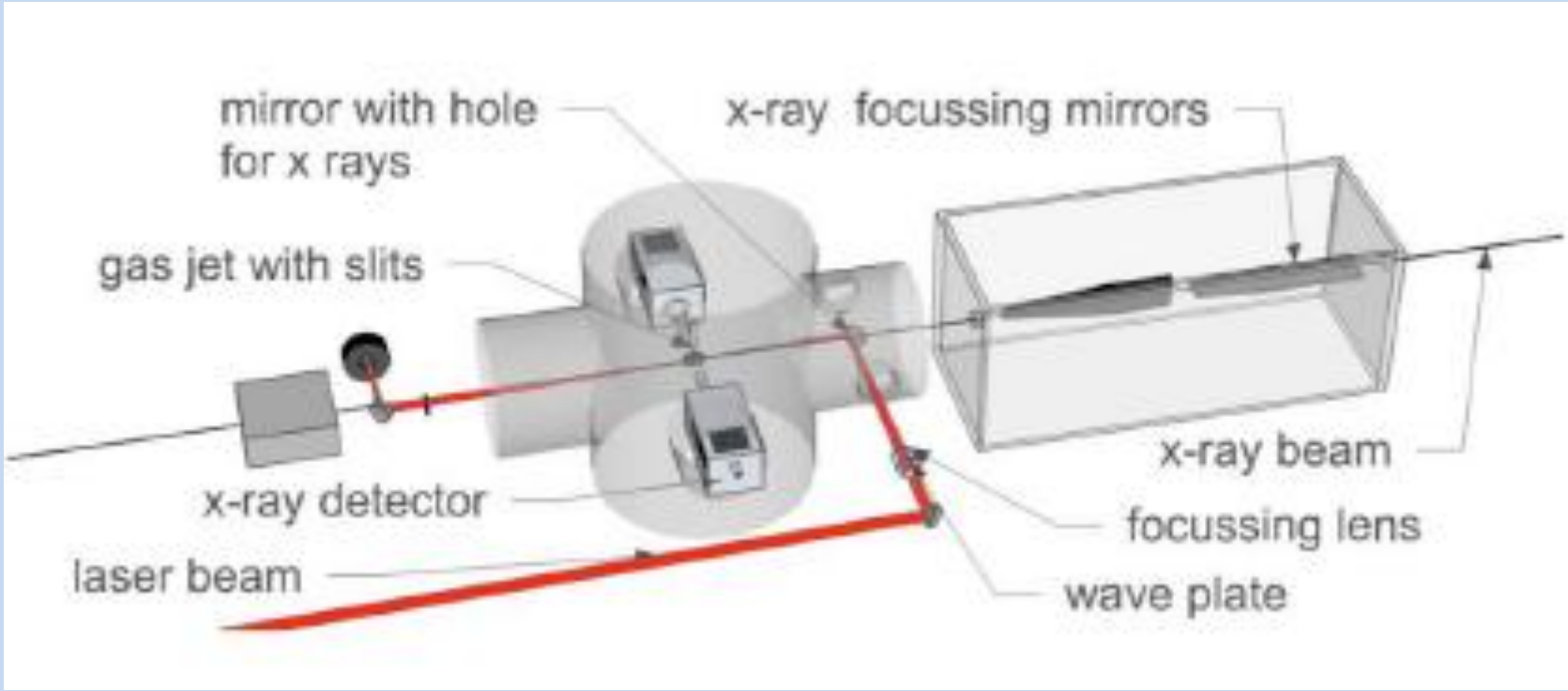
Pump-Probe Correlation: Fluorescence of Ionized Gas

- A laser pulse ionizes Kr to Kr^+, which opens a new, distinct fluorescence channel



Krassig, et al 2009, APS

Pump-Probe Correlation: Fluorescence of Ionized Gas



Krassig, et al 2009, APS

Pump-Probe Correlation: Fluorescence of Ionized Gas

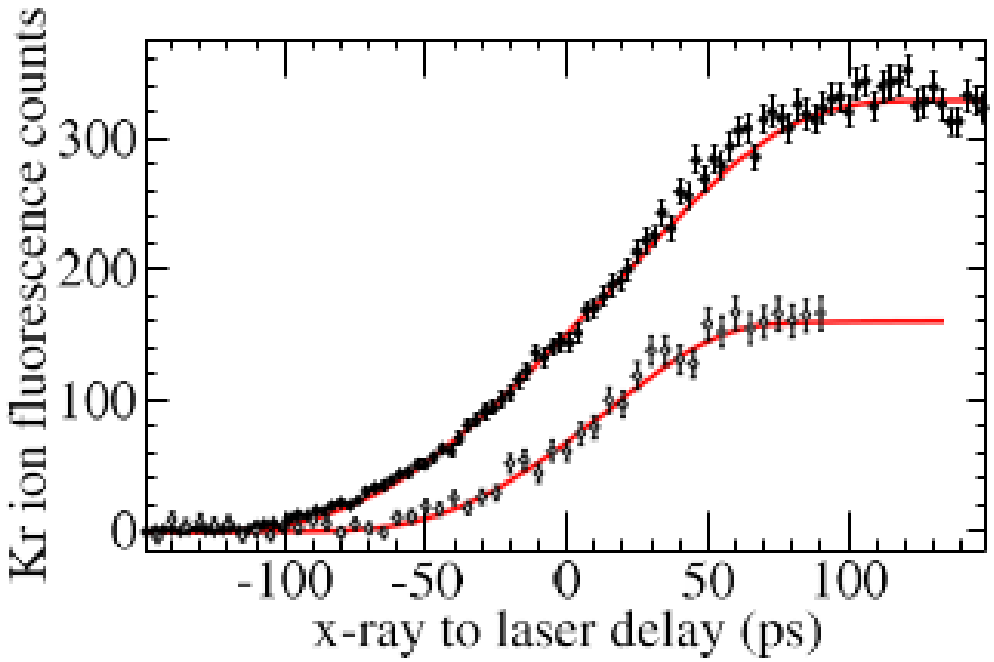


FIG. 3. (Color online) Cross-correlation data for hybrid fill (solid circles, 10 s/ch) and 24-bunch mode (open circles, 20 s/ch). A background from neutrals ($\sim$10) was subtracted. The solid curves are the cumulative integral of the respective x-ray pulse profiles from streak camera measurements.

Krassig, et al 2009, APS

Pump-Probe Correlation: Fluorescence of Ionized Gas

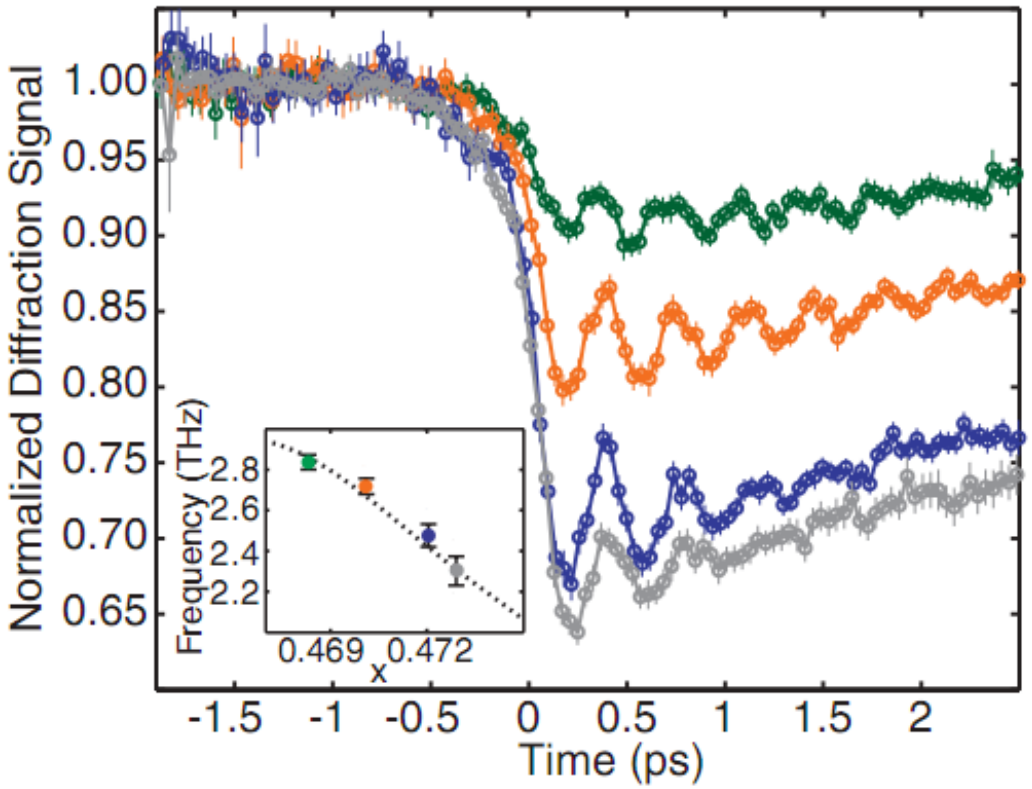
- This ionization takes only tens of fs and lasts for tens of ns, more than enough time to clock the ~100 ps x-ray pulse
- The main limiting factor is the jitter in the pulse frequency, ~5 ps
- This technique could be applied to FEL pulses of hundreds of fs.
- Limited to resonance energies of the gas

Example: Bismuth

- Pump-probe experiment at the Sub-Picosecond Pulse Source at SLAC
- 50nm thin film of bismuth hit with 70fs IR laser pulse to excite electrons, which modifies the interatomic potential
- A 100fs x-ray pulse probes the response of the lattice at the (111) Bragg peak.

Example: Bismuth

Fig. 1. Bismuth (111) x-ray diffraction efficiency as a function of time delay between the optical excitation pulse and x-ray probe for excitation fluences of 0.7 (green), 1.2 (red), 1.7 (blue), and 2.3 mJ/cm^2 (gray). The zero-delay point was set at the half maximum of the initial transient drop. The inset displays the optical phonon frequency as a function of the normalized atomic equilibrium position along the body diagonal of the unit cell *x* as measured by x-ray diffraction. The dotted curve represents the theoretical prediction obtained from DFT calculations of the excited-state potential-energy surface (*10*).



After ~20 ps, the Bragg angle began to shift, a result of strain stretching the unit cell

Fritz, Reis, et al 2007, SPPS

Example: Ga-As

- Pump-probe experiment at APS
- 1.5 µm thick (001) $Al_{0.3}Ga_{0.7}As$ layer grown on (001) bulk GaAs
- X-rays can resolve the alloy Bragg peak from the bulk peak
- The film is transparent to an IR laser, which, when absorbed by the bulk, generates a phonon which propagates through the film

Example: Ga-As

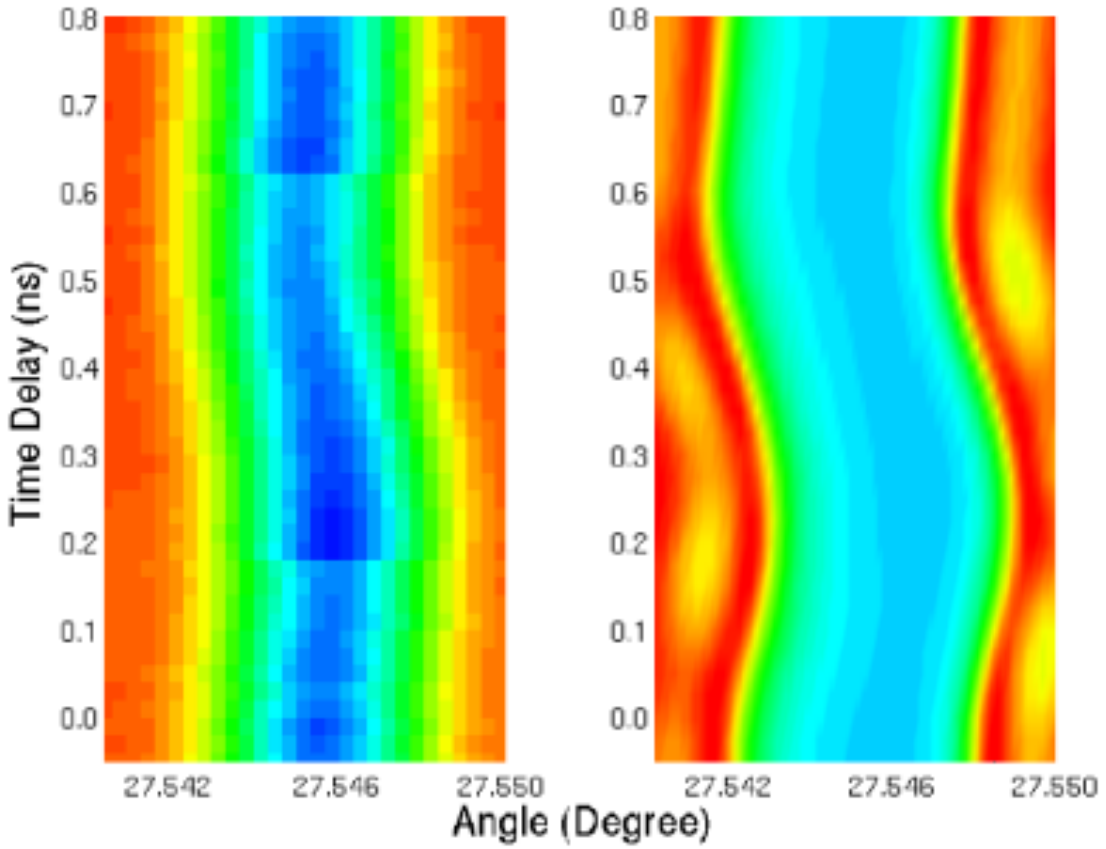


FIG. 4 (color online). Time and angle resolved x-ray diffraction from the $Al_{0.3}Ga_{0.7}As$ film showing the propagation of a short acoustic pulse that was generated in the GaAs substrate. Experimental data are on the left and a simulation in which strain is incorporated into dynamical diffraction theory is on the right.

References

- Ultrafast X-Ray Scattering in Solids; D. A. Reis and A. M. Lindenberg, Topics Appl. Physics **108**, 371–422 (2007)
- A simple cross-correlation technique between infrared and hard x-ray pulses; Bertold Krassig, R. W. Dunford, et al, Appl. Phys. Lett. **94**, 171113 (2009), DOI:10.1063/1.3125256
- Imaging Atomic Structure and Dynamics with Ultrafast X-ray Scattering; K. J. Gaffney, et al, Science **316**, 1444 (2007), DOI: 10.1126/science.1135923
- Femtosecond Undulator Radiation from Sliced Electron Bunches; Khan, Holldack, et al, Phys. Rev. Lett. **97**, 074801 (2006)
- Clocking Femtosecond X-Rays; Cavalieri, et al, Phys. Rev. Lett. **94**, 114801 (2005)
- Generation and Propagation of a Picosecond Acoustic Pulse at a Buried Interface: Time-Resolved X-Ray Diffraction Measurements, Lee, Cavalieri, Reis, et al, Phys. Rev. Lett. **95**, 246104 (2005)
- Ultrafast Bond Softening in Bismuth: Mapping a Solid's Interatomic Potential with X-rays , Fritz, Reis, et al, Science, **315**, 633 (2007)
- https://slacportal.slac.stanford.edu/sites/lclscore_public/Lists/LCLS_FAQ/FAQ.aspx
- http://www.aps.anl.gov/Facility/Storage_Ring_Parameters/

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